NEUTRON TRANSMUTATION DOPING IN HANARO REACTOR

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1. INTRODUCTION

Neutron Transmutation Doping (NTD) of silicon is a method for the n-type silicon semiconductor production using nuclear reactions. The NTD of silicon is based on a nuclear transmutation where a silicon atom absorbing a neutron is replaced into a phosphorus atom and it has a role as a dopant for the n-type silicon. The amount of phosphorus atoms occurred determines a resistivity value which is a measure of property for the semiconductor material. With the NTD technique, it is possible to obtain much higher uniformity of dopant distribution in the bulk silicon crystal comparing with other conventional chemical doping methods [1, 2]. For a long time, the silicon semiconductors have been used in various applications such as integrated circuits, thyristors, transistors, etc., and especially a higher uniformity of the dopant distribution is required for higher power operating devices such as IGBT[1], IGCT[2], GTO[3], etc., in order to ensure the devices against a hot spot formations and a possible break down. Recently the demand for such power devices is increasing rapidly by the rapid increasing needs for green energy techniques and the energy saving techniques for various industrial applications, and now NTD is in the spotlight as a valuable topic in the commercial utilizations using the research reactors.

We have been conducting commercial NTD service since 2003 using two vertical irradiation holes at HANARO. We started with 5 inch NTD-Si production at an irradiation hole (NTD-2) and added 6 inch from 2005 using the same hole. From 2009 we started the irradiation service for 8 inch silicon crystal using the other irradiation hole (NTD-1). In 2009, a total of 18 tonnes NTD-Si including 4 tonnes of 8-inch was supplied to the wafer companies of the world. At present, 5 and 6 inches are the main items of the NTD-Si market and the world demand is estimated to be 150~200 tonnes per year according to the market analysis form our customers. The 8 inch NTD-Si business is just in its infancy but it is expected to be a leading item with 6 inch in the market due to the growth of hybrid and electric cars before long.

This paper describes some special features and the present status of the NTD-Si production in HANARO of KAERI.

2. DESIGN CHARACTERISTICS OF NTD EQUIPMENT OF HANARO

2.1 Basic requirements

The silicon crystal may be the biggest one as a neutron irradiation target of the research reactors. In the NTD-Si production, the most important factor is to give a very uniform neutron dose all over a bulk crystal so that it has a highly uniform resistivity distribution after neutron irradiation. Note that the resistivity is inversely proportional to the total number of

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[1] IGBT : Insulated gate bipolar transistor
[2] IGCT : Integrated gate commutated thyristor
[3] GTO : Gate turn off thyristor

$Si^{30}(n,\gamma)\ Si^{31}$ reactions occurred inside the crystal, and the number of reactions is proportional to the total neutron dose to the crystal.

2.1.1 Radial uniformity

Because a silicon crystal has a large diameter, the neutron flux is different at everywhere along the periphery of the crystal. The most effective way for a uniform neutron irradiation in the radial direction is to rotate the crystal on its central axis continuously while it is irradiated. Another factor impacts on the radial uniformity is the neutron attenuation by silicon itself. Although silicon is very transparent to the neutrons and a neutron can move a long distance before it is removed by silicon atom, but the radial non-uniformity due to the neutron attenuation is unavoidable problem and becomes more serious for the crystal with larger diameter. Usually the market requirement for the radial uniformity is less than 5% for the 5 and 6 inch crystals and less than 8% for the 8 inch crystal. It is generally believed that more than 8 inch silicon crystal is not applicable to NTD, but studies are under progressing [3].

2.1.2 Axial uniformity

In usual research reactors, the irradiation holes have non uniform neutron distribution along its axial directional. In order to maximize the length of the silicon crystal per irradiation, an artificial modification of the original neutron distribution may be needed. Otherwise only small section of the hole can be practically used for high quality NTD-Si production. Actually, this is the most important factor that should be carefully considered for the successful NTD operation at the reactor site. The market required axial deviation of the final resistivity is usually within ±5~6% depending on the crystal diameter.

Practically three kinds of methods are used for the axially uniform neutron irradiation depending on the condition of each reactor: 1) travelling method as at BR2 of Belgium [4] or at RISO of Demark [5], 2) two times irradiation method in JRR-3M of Japan [6], and 3) neutron screening method as in OPAL of Australia .

2.1.3 Accuracy of neutron dose

Every silicon crystal should be so irradiated that it has the exact neutron dose pre-determined according to its target resistivity. The accuracy of irradiation is also very important as well as the uniformity of irradiation. In order to get an accurate irradiation, the real time neutron detector such as SPND[4] is usually used. During the irradiation, it gives the time accumulated neutron flux (dose) in real time and tells when the irradiation has to be finished. It cannot represent the exact neutron dose inside the silicon ingots however, sometimes the activation neutron detectors are uses together for the absolute measurements of the total neutron dose in the crystals.

2.2 NTD of HANARO

2.2.1 Irradiation holes for NTD

HANARO has two vertical irradiation holes for NTD application in the heavy water reflector tank surrounding the reactor core as shown in Figure 1. They are around 67 cm distant from the core center in the opposite directions and the inner diameters are 22 cm (NTD1 hole) and

[4] SPND: Self powered neutron detector

18 cm (NTD2 hole) respectively. The designed maximum thermal neutron flux is 5×10^{13} $cm^{-2}s^{-1}$ in both holes, while the fast neutron flux and gamma radiation level are very low enough to prevent the silicon crystal from defect by such radiation. The total length of a hole is 120, but the effective length is 70 cm.

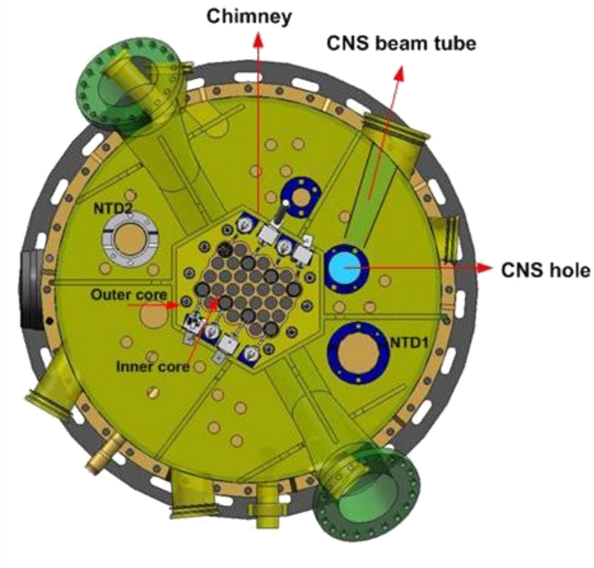


Fig. 1. NTD holes in HANARO.

2.2.2 Irradiation rig and neutron screen

We decide to use the neutron screening method considering every condition of HANARO and also productivity. With other two methods above mentioned, although they have advantages of simple design and easy manufacturing of necessary equipments, the maximum crystal length per irradiation is limited to less than 30 cm and more time and more hands are needed as well.

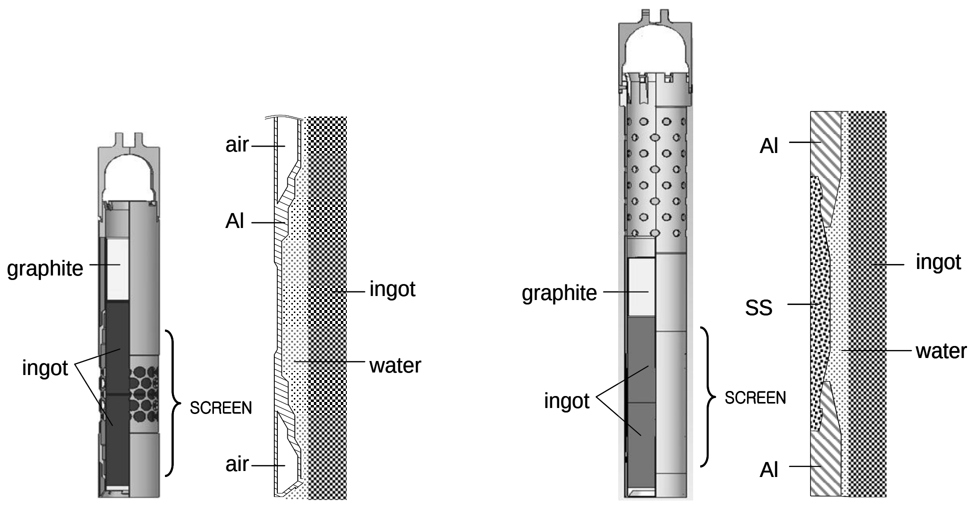
Nickel, titanium or stainless steel is used as the screen materials in usual applications but they are disadvantageous to the neutron economy due to their strong neutron absorbing power. These materials were completely excluded from the first stage of our feasibility study for NTD. Instead, it was confirmed that a neutron screen with a high performance can be achieved using mainly water and aluminum and it can give a great advantage of maximizing the neutron flux to the silicon because a little neutrons will be filtered through the screen [7]. As well, it was favorable for managing the radio wastes because aluminum does not create the long-lived radioactive isotopes.

Our study was also focused on the idea that a neutron screen does not necessarily need to be installed in the irradiation hole separately instead, it would be better to be integrated into the irradiation rig (a container of the silicon crystals). It was because that the axial neutron profile in the irradiation hole is going to be changed as time passes due to fuel burn effect. This effect is so small to be ignorable in usual cases but it was confirmed though our study that it can give more than ±5% change in the irradiation results. By doing that, it is possible to move screen position for optimized irradiation. Accordingly, we designed an irradiation rig made by only aluminum, of which wall thickness varies along its height. Gap water between the silicon crystal and the rig acts a dominant neutron filtering material in this case. In order to maximize the axial region of a flat neutron distribution and at the same time to maximize the neutron flux, it was necessary to filter neutron out a little at the central region of a rig and to raise neutron density at both ends. This can be achieved by making air pockets at the top and bottom sides of a rig wall and by putting additional cylindrical graphite blocks as neutron reflectors on the top and bottom of the crystals during the irradiation.

This concept was adopted in developing our first 5 inch irradiation rig for NTD2 hole as shown in Figure 2(a). The minimum thickness of the aluminum wall is 5 mm at the middle part and maximum thickness is 17.9 mm at the upper and lower part. While an upper graphite block is loaded into the irradiation rig following the silicon crystals, the lower graphite is installed in the irradiation hole permanently. The combination of a rig and two graphite reflectors provides a flat neutron distribution up to 605 mm long and also it provides a very high neutron flux more than 3.5×10^{13} $cm^{-2}s^{-1}$.

In case of irradiation rig for 6 inch ingots, it was impossible to make a desirable neutron screen with only aluminum and water because a very small gap space remains due to increased crystal diameter. Therefore, a substitute material of higher neutron absorbing power was needed and stainless steel was selected through the analysis of the several candidate materials. Figure 2(b) shows an optimized neutron screen for 6 inch crystals. Stainless steel is adopted only at the middle section of the rig and upper and lower sections are made by aluminum again. The graphite reflectors are included too. The rig has wall thickness varying from 2.2 to 5.1 mm in the screen section and provides a wide region of flat neutron distribution and a high neutron flux as like as 5 inch rig. Afterward we developed two more rigs for 6 and 8 inch silicon crystals for the irradiation at NTD1 hole. The design concept of them is similar to the previous rigs.

In the simulation using MCNP computer code, all irradiation rigs were expected to give an excellent axial uniformity within ±2.0% deviation [7, 8]. The simulation results for 5 and 6 inch are described in Figure 3 that is rated with the normalized total reaction rate of Si-30(n,γ)Si-31. From the test irradiations and also many times practical irradiations, it is confirmed that the axial deviation is within ±5.0% over 600 mm long for all kinds of ingots, and the RRG[5] is within 5 % for both 5 and 6 inch ingots and within 7% for 8 inch ingots [9,10]. The summary of each irradiation rigs is described in Table 1.



(a) 5 inch rig *(b) 6 inch rig*

Fig. 2. Irradiation rigs and neutron screens for NTD2 hole.

5 RRG (Radial resistivity gradient): A measure of radial uniformity of the silicon ingots

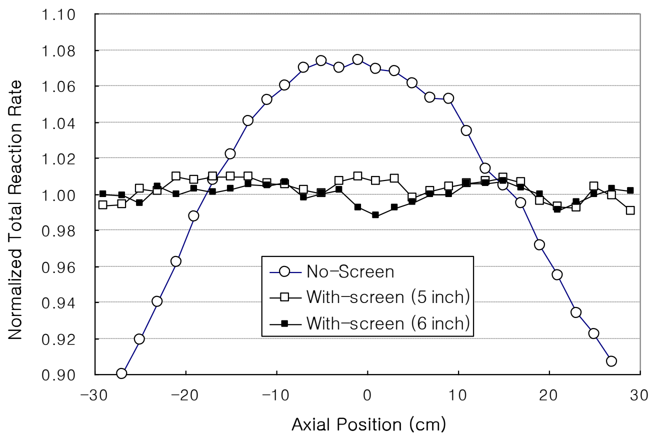


Fig. 3. Comparison of $Si^{30}(n,\gamma)Si^{31}$ reaction rate.

TABLE 1. CHARACTERISTICS OF IRRADIATION RIG FOR NTD IN HANARO

(unit : mm)		NTD2		NTD1	
		5 inch	6 inch	6 inch	8 inch
Hole	Inner dia.	180		220	
Screen	Outer dia.	166.0	→	215.0	→
	Inner dia.	130.2–156.0	155.8–161.6	156.4–176.2	207.0–210.0
	Thickness	5.0–17.9	2.2–5.1	19.5–29.4	2.5–203.1
	Material	Al, H_2O, Air	Al, SS, H_2O	Al, H_2O, Air	Al, SS, H_2O
Si ingot	Length	580–605	→	→	→
Neutron flux with Si ($cm^{-2}s^{-1}$)		~3.8×10^{13}	~3.5×10^{13}	~3.9×10^{13}	~3.6×10^{13}

3. IRRADIATION CAPACITY AND PERFORMANCE

The NTD holes of HANARO provide very high thermal neutron flux in spite of the neutron screen. The neutron flux is around 3.6×10^{13}~3.9×10^{13} $cm^{-2}s^{-1}$ at NTD1 and around 3.5×10^{13}~3.8×10^{13} $cm^{-2}s^{-1}$ at depending on the crystal diameter. The usual resistivity demanded from the market is in the range of 20 to 1000 Ω·cm. Then it takes about 8~10 hours to make 20 Ω·cm resistivity and takes less than 10 minutes for 1000 Ω·cm. The typical length of an irradiation batch of silicon crystals is 600 mm. Based on these factors and considering of HANARO's annual operating time (200 days per year), the production capacity of NTD-Si at HANARO is 50 tonnes per year including 20 tonnes of 5 and 6 inch using NTD2 and 30 tonnes of 6 and 8 inch using NTD1. It is a reasonably practicable prediction if inferred from the recent results.

After we started the first NTD irradiation service, HANARO had a long-term shutdown every year from 2005 until 2009 due to the installation and the initial commissioning of new facilities such as a high-pressure and high-temperature fuel test loop (FTL) and a cold neutron research facility (CNRF). Accordingly, HANARO was operated at a rate of 50~60% of its nominal operation capacity and NTD operation was the same. Fig. 4 shows an annual record for NTD-Si production with the operating ratio of HANARO. In spite of low operating rate before 2009, total production of NTD-Si was increased as the market demand was increased steadily. Currently, the NTD2 hole is operating at 100% capacity for 5 and 6 inch NTD-Si production. Only a few companies are producing 8 inch crystals for NTD and it would be take some more time before mass production.

In 2009, we provided a total of 18 tonnes NTD-Si including all marketable sizes and accomplished around 16 tonnes for the second quarter of 2010. We are expecting that the total production in 2010 will be over 30 tonnes because HANARO is planning 100% operating in this year.

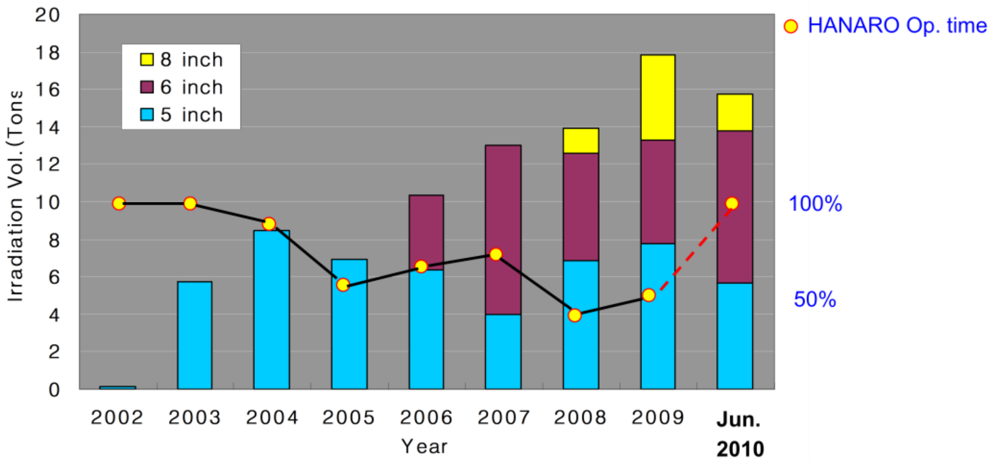


Fig. 4. Annual records of NTD-Si production.

Our own unique design of the irradiation equipments and the operation based on ISO quality management system enable the best quality NTD-Si production as well as world top productivity. The quality evaluation of all products in 2009 is made using the finally measured resistivity data by our customers. The irradiation accuracy and uniformity in axial and radial directions are indicated in Figure 5, Figure 6 and Figure 7 respectively. As shown in Figure 5, 90% of all irradiations are within ±3% in accuracy and 98% are within the customer's requirement (±5%). Only a few cases are deviated out of ±5%, but almost all of them were special cases with non-suitable conditions of dimension or initial resistivity distribution. Fig. 6 shows the axial uniformity distribution of 5 and 6 inch together. Excepting a couple of cases, the deviation of axial resistivity change is less than customer's requirement (5%) and the average deviation is around 1.5%. For the radial uniformity, it is out of our control but depends on initial condition of the irradiation batch in general. As shown in Figure 7, the RRG of 6 inch ingots is slightly higher than 5 inch but there is no big difference between 5 and 6 inch silicon crystals. For 8 inch crystals, the RRG is ranked 4~7%.

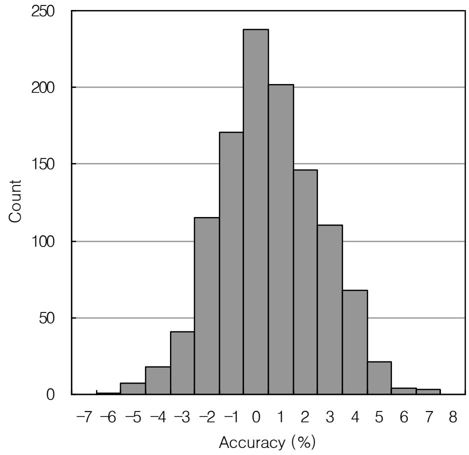


Fig. 5. Irradiation accuracy (2009)

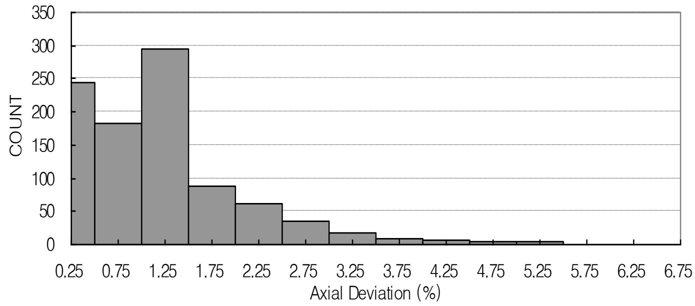
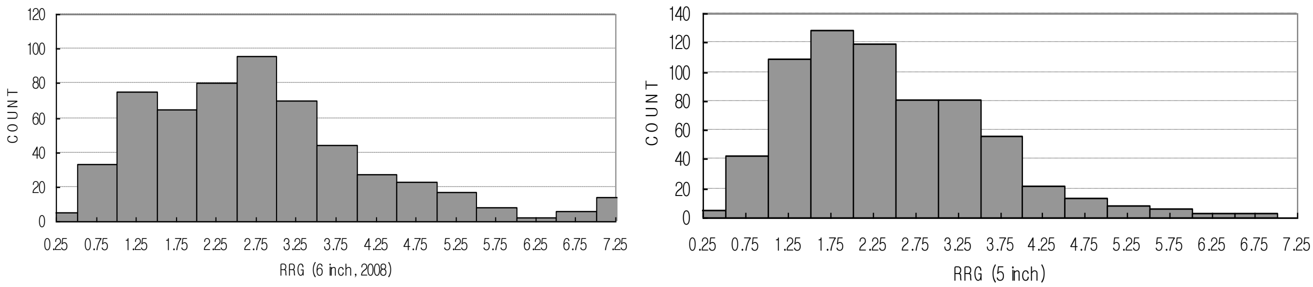


Fig. 6. Axial uniformity (2009).



(a) RRG for 5 inch *(b) RRG for 6 inch*

Fig. 7. Radial resistivity gradient (RRG) (2009).

4. MARKET SITUATION OF NTD-SI

Power devices are the semiconductor devices used as switches or rectifiers in power electronic circuits. The homogeneously doped semiconductor is a fundamental requirement such devices to overcome hot spot formations or break down due to the high operating voltage or current. However NTD of silicon is the only way to produce such semiconductor in a large scale because it is continuous problem to dope a bulk crystal with high uniformity.

These power devices are widely used in the various fields of industry such as long distance electric transmission, high-speed railway, air conditioner and industrial robots, etc. Recently worldwide attention to solving energy starvation and global warming is higher than ever and the rapidly increasing demand for hybrid and electric vehicles can be regarded in the same point of view. As a result, the demand for the power devices is also increased very rapidly every year. By a research report of a Japanese company the world market size for power devices is estimated at 120 billion US$ and will come to 160 million US$ in 2015. In case of IGBT where NTD-Si is used for mainly, it takes around only a few % of the total power devices but it will grow up more rapidly as IGBT module for hybrid and electric vehicles is expected to lead the IGBT market. The current NTD-Si demand is estimated at around 150~200 tonnes per year.

At present, the market is asking for more powerful devices with faster speed, lower electric loss and smaller size as well as applicability of higher electric power, however silicon devices have been faced with the a problem that they generate high heat due to increasing on-resistance when their internal voltage is increased. Because it comes from the material properties of silicon itself, other substitute materials have been developed. Diamond, silicon carbide or nitride such as GaN have better material properties such as wider band-gap, higher breakdown voltage and faster drift speed of carrier and better heat conductivity. Theoretically the most appropriate semiconductor material for power device is diamond, but it can't be practical because of the high cost and the difficulty in making into a large crystal. The alternative one is silicon carbide (SiC), which shows mid-range properties between silicon and diamond.

As silicon melts congruently at 1410°C of 1 atm, it is possible to have a stable growth of crystal with excellent crystalline and fast growing speed. In comparison, SiC undergoes a peritectic reaction[6] at 1 atm, therefore the liquid phase growth of SiC is very difficult. It is only possible under the condition of very high temperature over 3200°C and high pressure over 100 000 atm as like diamond synthesis. It is not realistic as of now and instead the sublimation phenomenon of SiC from gas state into solid state is mainly used for its crystallization. Currently only small size SiC crystal is available for the very restricted applications requiring very high frequency or very high power.

Another possibility is an epitaxial growth of the crystal. Epitaxial growth is a process of depositing a thin layer (around 0.5 to 30 microns) of single crystal material on the surface of other single crystal substrate. In this process, the substrate acts as a seed crystal and depositing material has the same structure and direction with the substrate. SiC substrate is also available using epitaxial growing as well as Si substrate. The epitaxial growing method is mainly utilized for the power device fabrication such as IGBT because of higher breakdown voltage across the collector-substrate junction and low collector resistance. However, it has a limitation that all substrates have to be produced in a wafer scale not a bulk crystal as silicon.

It seems to be true that SiC and epitaxy are the most possible competitors in the power device market. But it is also true that they have critical problems to be solved before they enter the market competition. No matter how the results of these new technologies turn on, it is not possibly realistic to expect that they will eventually replace all silicon based power devices for a while due to high cost and low productivity. On the contrary, NTD-Si can be produced in a large scale but it has a limitation in operating voltage. Therefore there may be a clearly differentiated market sharing between NTD-Si and SiC or other substrates.

[6] An isothermal, reversible reaction between two phases, a liquid and a solid

5. CONCLUSION

Worldwide attention to solving energy starvation and global warming is higher than ever. Changes for energy saving and high efficiency are progressing in every area of industry. Here, the use of power devices is one contributing factor. It is substituting the existing low power devices and especially the needs of power devices are soaring with the commercialization of hybrid and electric cars.

As of 2009, the use of NTD-Si is expected only 150~200 tonnes per year. This is because there are not many research reactors capable of mass production of NTD-Si and therefore the market size is determined by the supply capacity. At present, the only method to produce high semiconductor material for high power devices in large scale is the NTD using nuclear reactors. New technologies like SiC or epitaxi are being developed but NTD is much more competitive to consider production cost and productivity. Therefore there may be a clearly differentiated market sharing between NTD-Si and other high tech semiconductor materials and steady increasing of the NTD-Si market is expected too.

To meet the increase of semiconductor market and for the stable supply of NTD-Si, some research reactors are encouraging technology development. It is just as well that the NTD environment is reflected in a new research reactor design. Now NTD is becoming a very important area in the industrial use of nuclear research reactors. And more attention should be given to the NTD as a new business for a great added value to research reactor management

6. REFERENCES – JUST AN EXAMPLE!

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